Photoelectrochemical Water Splitting with TiO_2 Thin Films: Effects of Thickness, Porosity, and Illumination Configuration

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Nanostructured TiO_2 photoanodes have attracted great attention for solar fuel generation via photoelectrochemical water splitting. However, to date, their application remains limited due to an incomplete understanding of how morphology and thickness influence the separation, transport, and transfer of photogenerated charge carriers. In addition, little attention has been paid to the critical role of illumination direction, which determines the spatial distribution of charge carriers. In this sense, we conducted a systematic investigation into the synergistic impact of morphology (mesoporous vs dense), thickness (thin vs thick), and illumination configuration (back vs front) on the photoelectrochemical performance of nanostructured TiO_2 films prepared by dip-coating on FTO substrates. We performed a detailed structural, optical, photoelectrochemical, and ultrafast charge carrier dynamic characterization of each prepared photoanode. The photocurrent tests reveal that mesoporous photoanodes are globally less efficient than dense ones, regardless of thickness and illumination direction. Electrochemical and ultrafast techniques indicate that this is due to the increased density of surface defects, a weaker electric field, as well as slower charge transfer and transport, which promote higher recombination losses, mainly under front-side illumination conditions. These findings indicate that optimizing not only the illumination mode but also film thickness and morphology is essential to achieve high photoelectrocatalytic activity, and that crystallite size might have a more relevant role in photoassisted processes than surface area, which can introduce defects detrimental to performance.